

# **Aerosol-Cloud-Radiation Interactions in Atmospheric Forecast Models**

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## **LONG-TERM GOALS**

The long-term goal of this project is to gain a deep understanding of the role of atmospheric aerosols in affecting transmission of radiation through the atmosphere and in influencing cloud properties.

## **OBJECTIVES**

The scientific objectives of this project are to identify the specific manner in which atmospheric aerosols determine cloud properties and to represent these interactions in atmospheric models. The technological objectives are to develop state-of-the-art instruments for aircraft sampling of aerosols that advance the long-term goals of the project.

## **APPROACH**

The main technical approach is to conduct aircraft studies of the atmosphere, in which comprehensive sampling of atmospheric particles and radiative and cloud properties is carried out. The aircraft studies are complemented by laboratory investigations and theoretical analysis. Key individuals participating in this work are Professors John H. Seinfeld and Richard C. Flagan at the California Institute of Technology and Dr. Haf Jonsson at Naval Postgraduate School. Professor Seinfeld serves as Principal Investigator. Professor Flagan plays a key role in instrumentation development and planning of aircraft operations. As Chief Scientist of CIRPAS, Dr. Jonsson oversees all aspects of aircraft measurements and data management.

## **WORK COMPLETED**

The major work completed this year was: (1) analysis of data from the Marine Stratocumulus Experiment I (MASE I) carried out in Monterey, CA during July 2005; (2) execution of MASE II in Monterey, CA during July 2007.

## **RESULTS**

### **1. Marine Stratus/Stratocumulus Experiment**

The Marine Stratus/Stratocumulus Experiment I (MASE I) field campaign was undertaken during July 2005 off the coast of Monterey, California (Lu et al., 2007). The goal of MASE was to make

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state-of-the-art measurements of aerosols and clouds in the important regime of eastern Pacific marine stratocumulus clouds. The MASE experiment was undertaken to address following scientific questions: (1) Does cloud droplet number concentration increase in response to increasing subcloud aerosol amount? And, is cloud droplet size reduced? (2) Is the drizzle rate suppressed owing to more numerous cloud droplets? Furthermore, does the cloud liquid water path or averaged liquid water content increase because of drizzle inhibition? (3) Do we observe an aerosol-induced droplet spectral dispersion effect?

The cold ocean surface, in combination with warm, dry air aloft, induces the formation of one of the world's most persistent stratocumulus cloud decks. The region of the Pacific atmosphere adjacent to the coast of California experiences air masses of very clean, background air as well as ones having strong anthropogenic influences. In addition, the prevalence of shipping lanes in this region offers the opportunity to study directly the effect of significant localized aerosol perturbations from ship emissions on marine stratocumulus properties, so-called ship tracks. The eastern Pacific region adjacent to the coast of California is, therefore, an ideal test bed for studying aerosol-cloud interactions. The July time frame was selected because it is the month during which the coverage of stratocumulus in this region is at its maximum. A total of 13 science flights were conducted between 2–17 July. Measurements were carried out on board the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter aircraft during the mission. Of the 13 Twin Otter flights that were conducted during MASE, six encountered strong, localized perturbations in aerosol concentration, size, and composition consistent with ship emissions. The subcloud aerosol and its impact on the stratocumulus layer were analyzed using a detailed aircraft cloud profiling strategy. Clear effects of enhanced aerosol loading in these cases were observed in cloud droplet concentration and droplet size distributions.

Thirteen clouds sampled in the region 123.5–121.5°W and 35.75–36.75°N were selected for detailed analysis; subcloud aerosol number concentrations varied from 70 to 1300 cm<sup>-3</sup>, with some cases exhibiting ship tracks. Among the clouds sampled, that observed on July 5 was clearly impacted by a ship track, as confirmed by in situ aircraft measurements and GOES near-IR satellite imagery (see Figures 1 and 2). Multiple airborne horizontal traverses (~30 km) through the ship track and unperturbed regions provide insights into variations of cloud properties on a 50-m (~1 Hz) scale both horizontally and vertically. Comparison of ship track and clean regions in the upper portion of the cloud shows that the ship track region exhibited a smaller cloud drop effective radius, reduced drizzle drop number concentration, and larger cloud LWC than the adjacent clean regions. The ship track region also exhibits a smaller cloud drop spectral width and relative dispersion, in accord with LES predictions of Lu and Seinfeld (2006) based on meteorological conditions in the ASTEX and FIRE experiments. The ensemble average of aerosol and cloud conditions over the 13 cloud regions sampled were also computed. For these, as subcloud aerosol number concentration increases, cloud drop number concentration increases. The magnitudes of aerosol and cloud drop number concentrations and their dependences lie roughly in the range as those reported by Martin et al. (1994) for a variety of locations. Averaging over all 13 clouds, as  $N_a$  (or CDNC) increases, the cloud drop effective radius decreases. Drizzle was prevalent in virtually all the clouds sampled. Drizzle drops were found to be most numerous and smallest near cloud top. The data suggest that more polluted clouds have fewer embryonic drizzle drops near cloud top, resulting in a smaller cloud base drizzle rate. The smaller cloud base drizzle rate, however, does not result in a larger LWP when compared with the clean clouds. It appears that direct conversion of cloud drops to drizzle is insufficient to explain the dependence of LWP on aerosol number concentration; drizzle intensity and the dynamic adjustment of the cloud in

response to drizzle in-cloud latent heating, subcloud evaporative cooling, and cloud top entrainment would need to be taken into consideration (Ackerman et al., 2004; Lu and Seinfeld, 2005).

Averaged results over all 13 clouds show that more polluted clouds have narrower spectral width. The MASE data exhibit a clear positive correlation between cloud drop spectral dispersion  $d$  as a function of  $N_a$ . In general, on the ensemble cloud scale, increases in aerosol number concentration result in cloud LWP decreases and dispersion broadening. The contrast is due to the fact that, at the ensemble scale, clouds are influenced by different meteorological conditions that result in different cloud top entrainment, cloud base updraft variance, drizzle intensity, and surface evaporation; at the scale of a single cloud, the ship track and clean regions are embedded in the similar sounding profile, and the aerosol-induced changes are compared irrespective of the variations in LWP and dispersion due to ambient conditions.

## **2. The Source of Organic Acid Aerosol Layers above Clouds (Sorooshian et al., 2007)**

Organic carbon (OC) is a major component of atmospheric particulate matter. Organic acids constitute a significant fraction of particulate organic carbon. Primary emissions from fossil fuel combustion, biomass burning, and biogenic activity are sources of particulate oxalic acid; photooxidation of volatile organic compounds (VOCs), particularly aromatic hydrocarbons, followed by condensation onto preexisting aerosols is also a source. Blando and Turpin (2000) suggested that organic acids are likely particle-phase compounds formed by cloud and fog processing. Oxalic acid, which has also been shown to be formed by aqueous-phase chemistry in cloud droplets, remains in the aerosol phase after subsequent droplet evaporation. In the aqueous phase, oxalic acid is formed by oxidation of either glyoxylic acid or by the oxidative decay of malonic acid, which, in turn, is formed by the oxidation of higher order dicarboxylic acids. Sorooshian et al. (2006) showed that the glyoxylic acid oxidation pathway is significantly more efficient at producing oxalic acid, as compared to malonic acid oxidation. Aqueous-phase intermediates to glyoxylic acid include glyoxal, methylglyoxal, glycolic acid, pyruvic acid, and acetic acid.

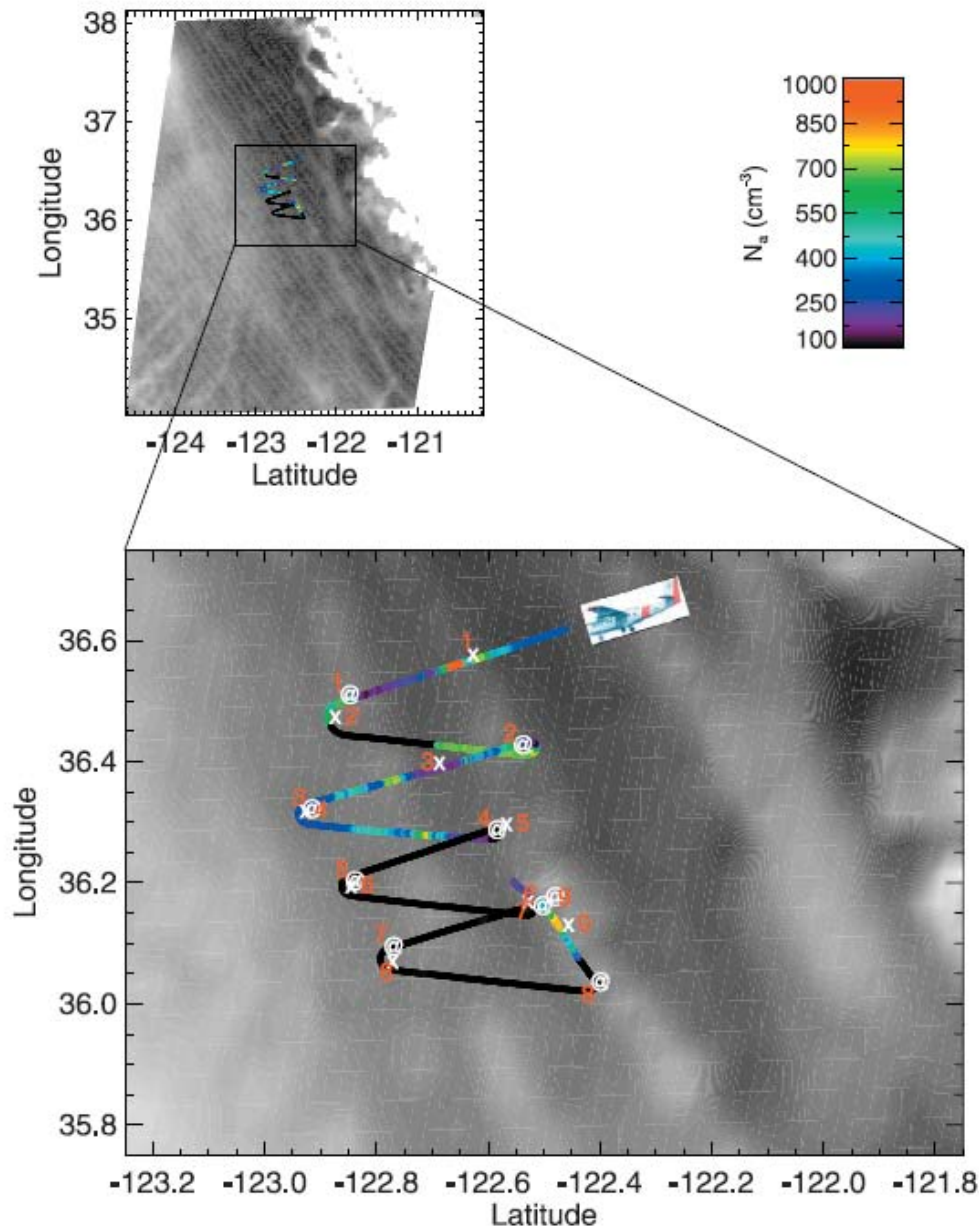
In July 2005, the Center for Interdisciplinary Remotely-Piloted Aircraft Studies (CIRPAS) Twin Otter (TO), based at Marina, CA, participated in the Marine Stratus/Stratocumulus Experiment (MASE I), focused on probing aerosols and stratocumulus clouds over the eastern Pacific Ocean off the coast of northern California (see 1 above). During August and September 2006, the CIRPAS TO also participated in the Gulf of Mexico Atmospheric Composition and Climate Study, in which 22 flights were devoted largely to probing aerosolcloud relationships over the Gulf of Mexico and the inland Houston area. In both MASE and GoMACCS, a ubiquitous layer of organic aerosol was found above cloud.

The goal of the present work is to evaluate, on the basis of the two field datasets, evidence and possible mechanisms for the existence of pervasive organic aerosol layers above clouds. We focus especially on organic acid levels in this work; particulate concentrations of organic acids and sulfate, and their ratios to each other, are compared above the boundary layer (BL) and below, inside, and above BL cumulus clouds. Results from large eddy simulations (LES) are used to provide support for a source of organic acid aerosols above cloud as residual particles from evaporated droplets.

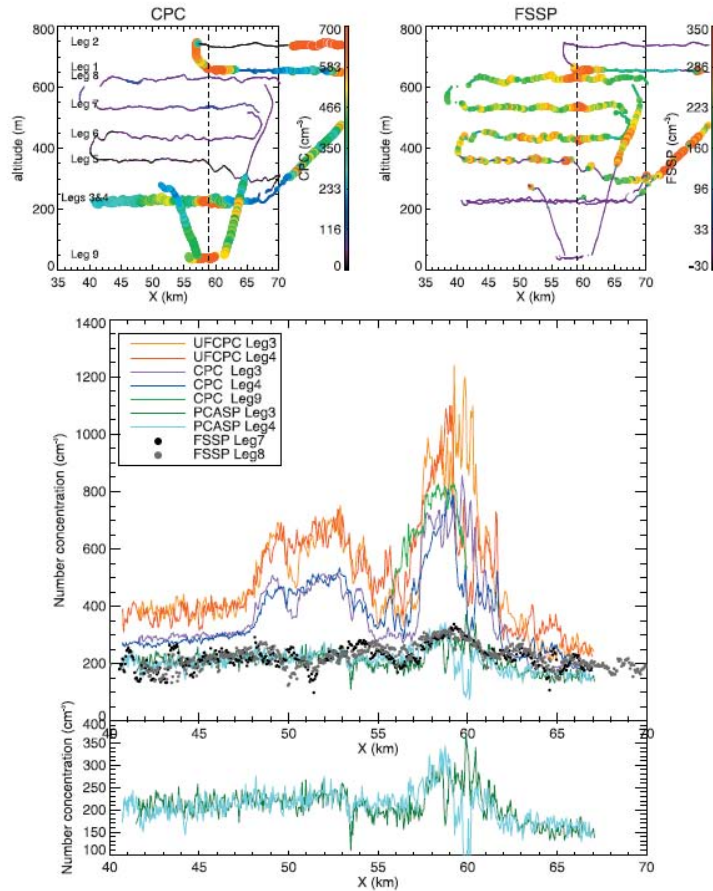
During MASE I, stratocumulus cloud bases typically ranged between 100 and 400 m, with tops between 350 and 700 m. Cumulus cloud bases during GoMACCS ranged between 500 and 800 m, with tops

between 800 and 3000 m. Typical stratocumulus liquid water content (LWC) values ranged between 0.1 and 0.5 g/m<sup>3</sup>, with a maximum of 0.7 g/m<sup>3</sup>, during MASE; cumulus LWC values between 0.1 and 1 g/m<sup>3</sup> were typically observed during GoMACCS, with a maximum of 2.1 g/m<sup>3</sup>. Sounding profiles up to cloud bases indicate that the BL tended to be well-mixed during both field campaigns. An estimate of the in-cloud residence time of an air parcel in a well-mixed BL can be obtained from the cloud volume fraction of the BL, values of which for both field studies were usually between 20 and 50%. The volume fraction was calculated by dividing the average cloud depth observed during cloud field legs by the BL height; uncertainties in this calculation include the assumption that there was a continuous cloud deck of the same depth everywhere in the BL and that there was no deviation in cloud thickness. For GoMACCS, this likely represents an upper bound since cloud fractions were on the order of 10%. Organic acids constituted a significant portion of the above-cloud aerosol mass during both campaigns; on average, organic acid mass above cloud during MASE and GoMACCS was  $0.07 \pm 0.04 \mu\text{g}/\text{m}^3$  ( $4.9 \pm 3.6\%$  of PILS mass) and  $0.14 \pm 0.14 \mu\text{g}/\text{m}^3$  ( $6.5 \pm 5.1\%$  of PILS mass), respectively.

An on-board particle-into-liquid sampler (PILS) quantified inorganic and organic acid species with  $\leq 5$ -min time resolution. Ubiquitous organic aerosol layers above cloud with enhanced organic acid levels were observed in both locations. The data suggest that aqueous phase reactions to produce organic acids, mainly oxalic acid, followed by droplet evaporation is a source of elevated organic acid aerosol levels above cloud. Oxalic acid is observed to be produced more efficiently relative to sulfate as the cloud liquid water content increases, corresponding to larger and less acidic droplets. As derived from large eddy simulations of stratocumulus under the conditions of MASE, both Lagrangian trajectory analysis and diurnal cloudtop evolution provide evidence that a significant fraction of the aerosol mass concentration above cloud can be accounted for by evaporated droplet residual particles. Methanesulfonate data suggest that entrainment of free tropospheric aerosol can also be a source of organic acids above boundary layer clouds.



**Figure 1.** GOES-10 satellite Band 2 (centered at  $3.9 \mu\text{m}$ ) satellite picture at 17:45 UTC, 5 July 2005. The horizontal resolution of this band is 4 km. GOES data are obtained from the NOAA Satellite and Information Service. Flight path is colored according to aerosol number concentration measured by CPC. Horizontal legs are marked on the path (x, start: @ end). Because liquid water at this wavelength is moderately absorbing, sunlight does not penetrate the cloud deeper than about 100 m, so that the cloud albedo depends mainly on the cloud droplet size in the upper region of the cloud. The near-IR imagery reveals two marked brighter (high reflectance) narrow, curvilinear cloud lines in the sampling period. Between and outside the two brighter ship track impacted clouds are darker regions that are still cloudy from the visible channel picture. The ship track on the west was sampled with nine horizontal traverses, seven through the cloud layers and two above and below clouds, respectively. The horizontal flight legs are numbered based on the time sequence.



**Figure 2.** *Flight paths colored according to aerosol number concentration (upper left) and cloud drop number concentration (upper right). The flight path has been shifted horizontally by the wind drift so that the vertical profiles can be aligned. The lower panel shows the below or near cloud base aerosol number concentration from the Passive Cavity Aerosol Spectrometer Probe (PCASP), CPC, and UFCPC, and cloud droplet number concentration in the middle to upper regions of the stratocumulus cloud from FSSP. The PCASP data are masked by the FSSP data so as to be separately plotted at the bottom. A noticeable peak (orange to red color) in aerosol number concentration located around  $X = 59$  km is shown below and above cloud (Figure 4a), which is coherent with the apparent peak in cloud drop number concentration in the same location (Figure 4b). Figure 4c shows clearly that the horizontal distribution of cloud droplet number concentration varies with the horizontal distribution of below-cloud aerosol number concentration. Both Condensation Particle Counter (CPC) and Ultrafine CPC (UFCPC) data exhibit covariabilities with cloud droplet number concentration. The difference between data from the UFCPC and the CPC instruments represents the number concentration of ultrafine particles between 3 and 12 nm diameter; this indicates that freshly nucleated particles in the Aitken mode are significant in the major ship track region. Another smaller peak in CPC and UFCPC located around  $X = 48 - 54$  km is also seen at a similar  $X$  location in the cloud drop number concentration data. The spatially coherent peaks in aerosol and cloud number concentrations and the narrow curvilinear cloud lines on the satellite imagery suggest that increases of cloud droplet number concentration are associated with and can be ascribed to increases in below-cloud CCN concentration by particles emitted or formed from ship effluent.*



## REFERENCES

- Ackerman, A. S., M. P. Kirkpatrick, D. E. Stevens, and O. B. Toon (2004), The impact of humidity above stratiform clouds on indirect aerosol climate forcing, *Nature*, 432, 1014–1017.
- Blando, J. D.; and Turpin, B. J., (2000), Secondary organic aerosol formation in cloud and fog droplets: a literature evaluation of plausibility. *Atmos. Environ.*, 34, 1623-1632.
- Lu, M. L., and J. H. Seinfeld, (2006), Effect of aerosol number concentration on cloud droplet dispersion: A large-eddy simulation study and implications for aerosol indirect forcing, *J. Geophys. Res.*, 111, D02207, doi:10.1029/2005JD006419.
- Lu, M. L. and Seinfeld, J. H., (2005), Study of the Aerosol Indirect Effect by Large-Eddy Simulation of Marine Stratocumulus, *J. Atmos. Sci.*, 62, 3909-3932.
- Lu, M. L., W. C. Conant, H. H. Jonsson, V. Varutbangkul, R. C. Flagan, and J. H. Seinfeld, (2007), The Marine Stratus/Stratocumulus Experiment (MASE): Aerosol-cloud relationships in marine stratocumulus, *J. Geophys. Res.*, 112, D10209, doi:10.1029/2006JD007985.
- Martin, G. M., Johnson, D. W., and Spice, A., (1994), The measurement and parameterization of effective radius of droplets in warm stratocumulus clouds, *J. Atmos. Sci.*, 51, 1823-1842.
- Sorooshian, A.; Varutbangkul, V.; Brechtel, F. J.; Ervens, B.; Feingold, G.; Bahreini, R.; Murphy, S. M.; Holloway, J. S.; Atlas, E. L.; Buzorius, G.; Jonsson, H.; Flagan, R. C.; and Seinfeld, J. H., (2006), Oxalic acid in clear and cloudy atmospheres: Analysis of data from International Consortium for Atmospheric Research on Transport and Transformation 2004. *J. Geophys. Res.*, 111, D23S45, doi:10.1029/2005JD006880.
- Sorooshian, A.; Lu, M. L.; Brechtel, F. J.; Jonsson, H.; Feingold, G.; Flagan, R. C.; and Seinfeld, J. H., (2007), On the Source of Organic Acid Aerosol Layers above Clouds, *Environ. Sci. Technol.*, 41, 4647-4654.